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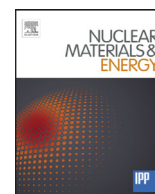
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Annealing effects on deuterium retention behavior in damaged tungsten



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ABSTRACT

Effects of annealing after/under iron (Fe) ion irradiation on deuterium (D) retention behavior in tungsten (W) were studied. The D₂ TDS spectra as a function of heating temperature for 0.1 dpa damaged W showed that the D retention was clearly decreased as the annealing temperature was increased. In particular, the desorption of D trapped by voids was largely reduced by annealing at 1173 K. The TEM observation indicated that the size of dislocation loops was clearly grown, and its density was decreased by the annealing above 573 K. After annealing at 1173 K, almost all the dislocation loops were recovered. The results of positron annihilation spectroscopy suggested that the density of vacancy-type defects such as voids, was decreased as the annealing temperature was increased, while its size was increased, indicating that the D retention was reduced by the recovery of the voids. Furthermore, it was found that the desorption temperature of D trapped by the voids for damaged W above 0.3 dpa was shifted toward higher temperature side. These results lead to a conclusion that the D retention behavior is controlled by defect density. The D retention in the samples annealed during irradiation was less than that annealed after irradiation. This result shows that defects would be quickly annihilated before stabilization by annealing during irradiation.

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1. Introduction

Tungsten (W) is a reference material of plasma facing materials (PFMs) in future fusion reactors [1,2] due to its good physical properties, such as lower sputtering rate and higher melting point. It is considered that W will be irradiated by 14 MeV neutron generated by D-T fusion reaction, which will introduce various types of defects like dislocation loop and vacancy [3]. It is well-known that the solubility of hydrogen isotopes for W is quite lower than that for graphite [1]. However, the radiation-induced defects may contribute to the retention enhancement of hydrogen isotopes in W, which refrains the development of effective fuel cycle in fusion reactors. In addition, the defects are recovered by annihilation and/or aggregated to form vacancy clusters under higher tempera-

ture during plasma operation [4], leading that the defect formation is greatly influenced by temperature. Therefore, it is important to understand the thermal annealing effects on deuterium retention for damaged W to estimate the hydrogen isotope retention.

To demonstrate the actual environment in fusion reactors, the best way is the use of fusion neutrons to introduce the damages. However, the production of radioactive materials by neutron irradiation will regulate the various surface analyses. The heavy ion irradiation method was used to introduce the damages in W [3–6].

This study was focused on the elucidation of defect behavior under higher temperature on D retention for damaged W. Therefore, two kinds of methods were adopted as follows. One is the irradiation at room temperature (R.T.) followed by annealing to evaluate the elementary step of defect recovery and aggregation to effect on D retention. The other is the irradiation at higher temperature to understand dynamic defect formation and recovery at the temperature environment closed to the actual fusion reactor. D was

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implanted in these samples with 1 keV D_2^+ , and thermal desorption spectroscopy (TDS) measurements were performed to evaluate the D retention behavior.

2. Experimental condition

Three disk-type polycrystalline W samples prepared from a W rod (purchased by A.L.M.T. Corp. Ltd.) under stress relieved conditions were used. These sample sizes were 10 and 6 mm in diameters and 0.5 mm in thickness using for TDS experiment and 3 mm in diameter and ~ 0.1 mm in thickness using for TEM observation. All the samples were mechanically polished to be the surface roughness of ~ 50 nm, cleaned in an ethanol ultrasonic bath and then heated at 1173 K in ultrahigh vacuum ($<10^{-6}$ Pa) for 30 min to remove the impurity gases and stress introduced by polishing. The surface morphology was described in our previous paper [6].

Thereafter, the samples were irradiated to introduce the defects by two different ways: (1) Post-annealed W: Irradiation at R.T. and applied annealing; (2) Dynamic-annealed W: Irradiation at elevated temperature. The damage concentration was calculated by SRIM code with the displacement energy of 50 eV [7].

- (1) 10 mm ϕ W samples were irradiated by 6 MeV Fe^{2+} at R.T. to damage levels in the range from 0.01 to 1.0 dpa using Takasaki Ion Accelerators for Advanced Radiation Application (TIARA) at JAEA. After irradiation samples were transferred to Shizuoka University and applied annealing at 573, 873 and 1173 K for 30 min in ultrahigh vacuum.
- (2) 6 mm ϕ samples were irradiated by 6.4 MeV Fe^{3+} up to the damage level of 0.1 dpa using Dual-Beam Facility for Energy Science and Technology (DuET) at 573, 873, and 1173 K

Then, these samples were implanted by 1 keV D_2^+ with an ion flux of $1.0 \times 10^{18} D^+ m^{-2} s^{-1}$ up to the ion fluence of $1.0 \times 10^{22} D^+ m^{-2}$ at R.T. TDS measurement was performed to evaluate D desorption behavior. The samples were heated up to 1173 K with a ramp up rate of $0.5 K s^{-1}$. In addition, positron annihilation spectroscopy (PAS) was performed to observe the vacancy-type defects formation for annealed samples in the damage level of 0.1 dpa using Mylar film including ^{22}Na as a positron source at Tohoku University. The defects formation and its thermal stability were observed for 0.3 mm ϕ W irradiated by TIARA in the damage level of 0.1 dpa by transmission electron microscope (TEM) (JEM 2000EX, JASCO Inc.) at Kyushu University. The samples were observed after annealing for 30 min in the temperature range from R.T. to 1173 K

3. Results and discussion

Fig. 1 shows the D_2 TDS spectra for 0.1 dpa damaged W samples with post-annealing at various annealing temperatures. The TDS spectra consist of three desorption peaks located at about 400, 600 and 800 K. According to previous studies [6,8,9], these desorption peaks are attributed to the desorption of D trapped by dislocation loops and/or adsorbed on the surface as Peak 1, that trapped by vacancies as Peak 2 and that trapped by voids as Peak 3, respectively. The D retention behavior is summarized in Fig. 2. Total D retention was clearly reduced as increasing annealing temperature, which indicates that the amount of D trapping sites was decreased by recovery of the defects. Therefore, the defect recovery and aggregation behavior were observed by TEM and PAS.

Fig. 3 shows TEM images for 0.1 dpa damaged W with post-annealing at various temperatures. Dense dislocation loops as black dots were formed by Fe^{2+} irradiation at R.T. and initiated to grow at 573 K. The dislocation size was increased and density was decreased with increasing temperature. Almost all the dislocation loops were recovered by annealing at 1173 K. On the other hands, D

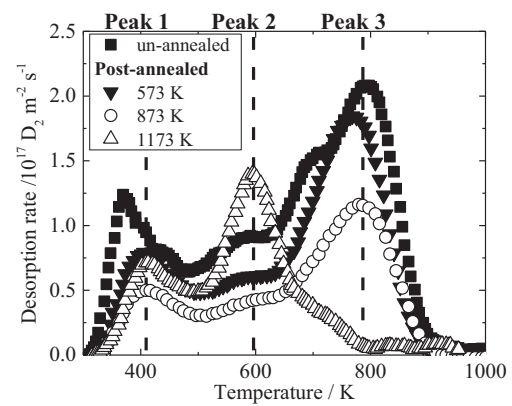


Fig. 1. D_2 TDS spectra for 0.1 dpa damaged W with post-annealing at various temperatures. The result for un-annealed W was also shown.

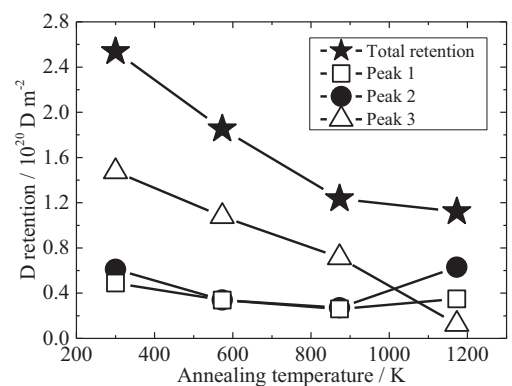


Fig. 2. Annealing temperature dependence on D retention at each desorption stage and total retention for post-annealed W.

desorption Peak 1 was not almost reduced even if annealing temperature was reached at 1173 K as shown in Fig. 1. It can be said that the amount of deuterium trapped by dislocation loops would be small. So, D retention is not controlled by the density of dislocation loops.

Fig. 4 shows the intensity and lifetime of long-lifetime positron in post-annealed W samples. The increases of intensity and lifetime indicate the growth of the void concentration and its size. The positron lifetime was increased with increasing annealing temperature, showing that the vacancy-type defects were aggregated to form vacancy clusters such as voids [10]. However, its intensity was reduced as the annealing temperature was changed from 573 K to 1173 K. This results show the voids were recovered by annealing at 1173 K, which is consistent with the D retention behavior, where the D retention as Peak 3 in Fig. 2 was clearly reduced. It can be said that the recovery of voids has a large impact on the reduction of D retention in damaged W.

D_2 TDS spectra for 0.01–1.0 dpa damaged W samples with post-annealing at 1173 K are shown in Fig. 5. D desorption behavior in the annealed W above 0.3 dpa was different from that in the 0.1 dpa damaged W sample. D retention as Peak 3 was almost disappeared by annealing at 1173 K for 0.1 dpa damaged W. However, that for 0.3 dpa damaged W was still remained. In addition, desorption temperature at Peak 3 was shifted toward higher temperature side with increasing the damage concentration, which was caused by that D trapped by trapping sites in heavily damaged zone undergoes trapping/detrapping processes [6]. These results suggest that the behavior of recovery and aggregation of the defects are controlled by damage concentration.

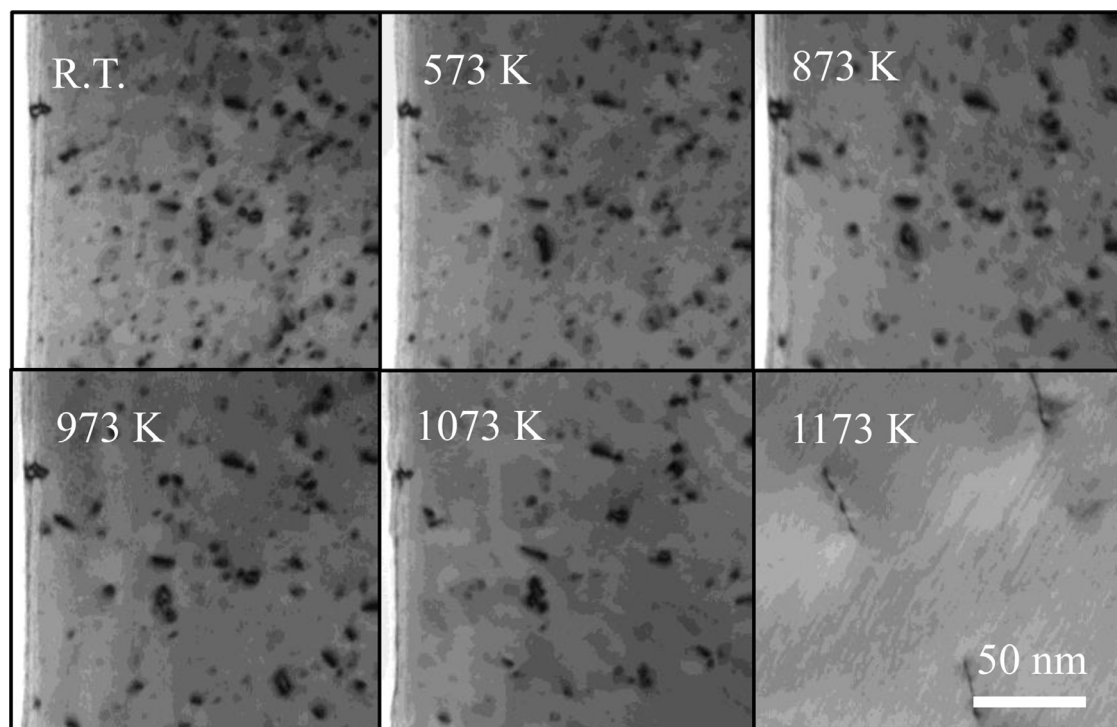


Fig. 3. TEM images (dislocation loops) for 0.1 dpa damaged W with post-annealing at various temperatures.

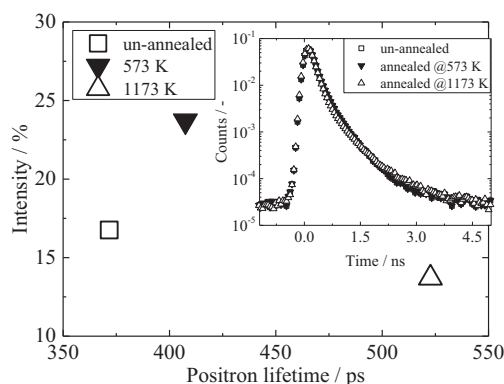


Fig. 4. Lifetime dependence of long-life positron in post-annealed W and positron annihilation lifetime spectra.

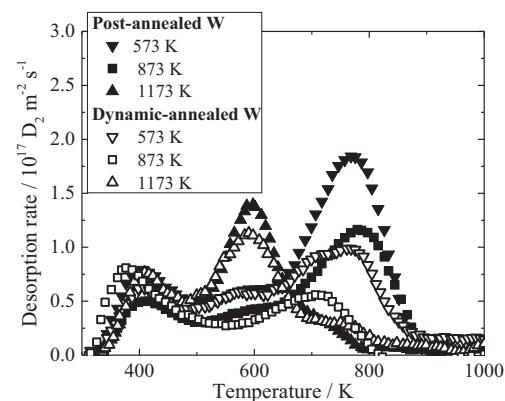


Fig. 6. Comparison of TDS spectra for dynamic-annealed W and post-annealed W.

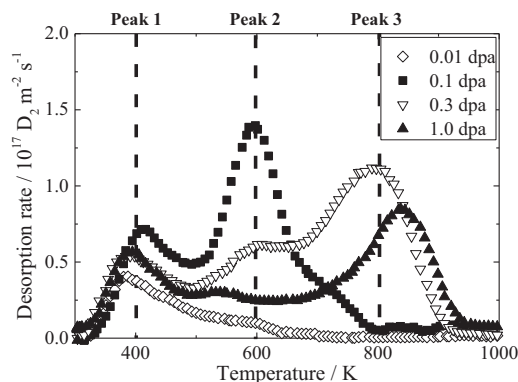


Fig. 5. TDS spectra for post-annealed W at 1173 K in the damage levels of 0.01–1.0 dpa.

Comparison of D desorption behavior for the dynamic-annealed W samples and post-annealed W samples is shown in Fig. 6. D retention as Peak 3 in the dynamic-annealed W was clearly smaller

than the post-annealing samples, and desorption temperature was shifted toward lower temperature side at lower annealing temperature (573, 873 K). On the other hand, no large difference was found in D desorption behavior in the dynamic-annealed and post-annealed W at 1173 K. It can be said that the recovery of defects was enhanced due to the increase of defect mobility by irradiation [4] and quick annihilation of trapping sites before stabilization to form clusters due to the formation and recovery of defects at the same time. However, at higher annealing temperature, thermal annealing effects would make a large contribution. Thus, D retention for dynamic-annealed samples is the same as that for post-annealed samples. To simulate these TDS spectra and evaluate the trapping energy, Hydrogen Isotope Diffusion and Trapping (HIDT) simulation code described in our previous paper in detail [6] was utilized. The parameters are shown in Table 1. Fig. 7(a) shows the simulation of D_2 TDS spectra. The trapping energies of Peaks 1–3 were calculated to be 0.65, 1.00 and 1.23–1.35 eV. It was conceivable that the broadening of D desorption Peak 3 should be attributed to the existence of several trapping states by changing of void sizes due to the recovery and aggregation. D depth profile

Table 1
Summary of parameters used in the HDT simulation code.

Variable	Value
Thickness	0.5 mm
Density of W atoms	$6.3 \times 10^{28} \text{ m}^{-3}$
Interstitial site density	$3.8 \times 10^{29} \text{ m}^{-3}$
Diffusivity: D_0	$2.9 \times 10^{-7} \text{ m}^2/\text{s}$
Diffusion energy: E_D	0.39 eV
Trapping energy 1	0.65 eV
Trapping energy 2	1.00 eV
Trapping energy 3	1.23–1.35 eV
Implantation depth for Fe ions	1.1 μm
Implantation depth for D ions	10 nm

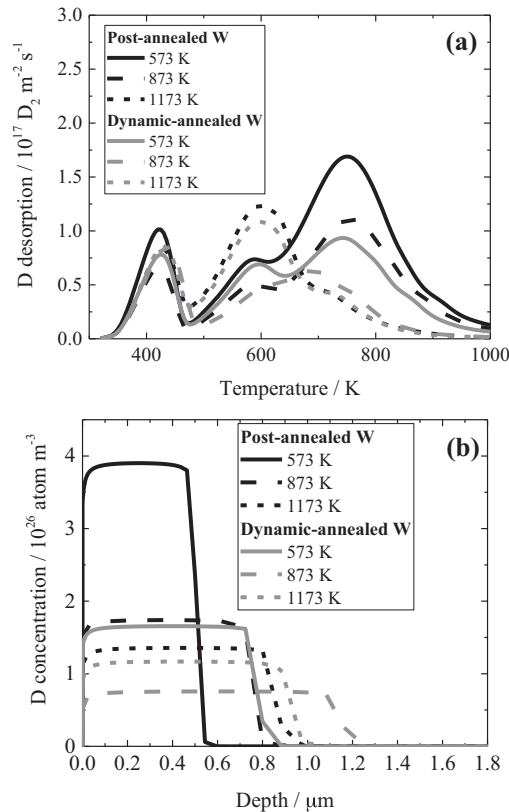


Fig. 7. (a) D_2 TDS simulation results and (b) D depth simulation results for Dynamic-annealed W and post-annealed W.

of the post-annealed W (Fig. 7(b)) showed that D for the dynamic-annealed W was penetrated into the deeper region than that for the post-annealed W. the defects in the dynamic-annealed W were easily recovered as compared to these in the post-annealed W as indicated above. Therefore, D was trapped by trapping sites at the deeper side easily because D trapped near surface region was decreased by the reduction of defect concentration due to the recovery of defects.

4. Conclusions

D retention behavior for post-annealed W and dynamic-annealed W were evaluated to elucidate the correlation between D retention and the defects recovery and aggregation behavior caused by thermal annealing in higher temperature environment during plasma operation. It was found that D retention was not controlled by the density of dislocation loops. The recovery of voids remarkably reduced D retention for 0.1 dpa damaged W. However, for the damaged W above 0.3 dpa, the voids were not recovered and they has grown, which controls the D retention. In addition, dynamic-annealing reduced D retention compared with that in post-annealed W at the same temperature with same damage concentration. It can be said that the damage concentration will control the D trapping site and its aggregation would sustain D retention in W. In addition, dynamic-annealing would reduce D trapping sites due to quick annihilation of the defects before stabilization. It can be concluded that control of dynamic temperature for plasma facing wall during fusion operation is one of keys to reduced the damage concentration and fuel retention. In addition, it is also important to evaluate the effects of impurity such as helium, and defect formation rate on the defect recovery behavior for demonstrate the fuel behavior.

Acknowledgments

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